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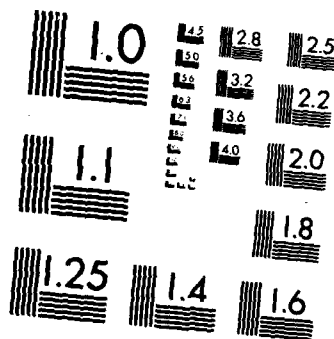
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**Gated Spectral Hole-Burning for Frequency Domain Optical Recording**

**by**

**W. Lenth and W. E. Moerner**

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Solid State Physics

## GATED SPECTRAL HOLE-BURNING FOR FREQUENCY DOMAIN OPTICAL RECORDING

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**ABSTRACT:** Materials exhibiting persistent spectral hole-burning via a gated mechanism are promising candidates for the development of frequency domain optical storage with storage densities beyond  $10^9$  bits/cm<sup>2</sup>. Gated hole-burning requires a secondary gating field for writing, permitting nondestructive reading in the absence of this field. Properties of gated hole-burning materials suited for a practical storage system are analyzed with particular attention to the required values of absorption cross section, density of centers, and effective hole-burning yield. The results permit evaluation of the usefulness of particular gated hole-burning materials for storage applications. Some general guidelines for photon-gated mechanisms using three-level and four-level systems are presented.

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## I. Introduction

The phenomenon of persistent spectral hole burning (PHB) permits use of the optical frequency for encoding digital data in a storage scheme called frequency domain optical storage [1] [2] [3] . A suitable storage material, which is kept at low (liquid helium) temperatures, contains optically active centers that exhibit an inhomogeneously broadened absorption line resulting from strain-induced frequency shifts of the atomic or molecular resonance. With a narrow-bandwidth tunable laser, specific frequency locations within the absorption line can be addressed by selective excitation of those centers that are resonant with the laser frequency. When the optical excitation is accompanied by a photochemical or photophysical process that induces a persistent population reduction of the selected centers, multiple spectral holes can be burned in the absorption line. The presence or absence of a spectral holes at given frequency locations can be used to represent binary data. The ratio of inhomogeneous to homogeneous linewidth,  $\Delta\omega_i/\Delta\omega_h$ , which can be as high as  $10^3 - 10^4$ , approximately determines the storage capacity of the frequency domain. Storage densities of  $10^9 - 10^{10}$  bits/cm<sup>2</sup> or even higher should be possible using tightly focused laser beams.

In most PHB materials investigated the photo-induced changes in the absorbing centers involve a single-photon process. Excessive photoreaction during the detection of spectral holes can only be avoided by using much lower light intensities than for hole burning. In optical storage applications, however, significant photo-induced bleaching during data interrogation would accumulate after a large number of reads and eventually the read signal-to-noise would degrade below tolerable levels. Recent modeling studies have addressed in detail the limited usefulness of single-photon hole burning mechanisms for a frequency-domain storage configuration[4]. However, non-destructive hole detection is possible if hole burning occurs via a gated mechanism that, in addition to optical excitation, requires a "gating" event to

initiate the reaction process leading to the formation of a persistent spectral hole. Such a gating event could be triggered by an electrical field, microwave radiation, or a second optical photon. Three experimental examples of gated spectral hole burning by two-step photoionization have been reported recently [5] [6] [7] . A first photon is used for frequency-selective excitation within the inhomogeneously broadened absorption line; the absorption of a second photon by the excited state results in photoionization and hole burning.

The general mechanism of gated PHB has been demonstrated but several materials properties await optimization before a practical storage system can be realized. The search for a suitable storage medium that fulfills all technological requirements presents a challenging task for solid-state materials research. The complex nature of gated PHB processes make a systematic approach for finding a satisfactory material rather difficult. In this paper we analyze desirable properties of gated PHB materials for optical storage applications.

## II. Analysis of Gated Hole-Burning Materials

In order to identify the critical properties of a suitable storage medium, one has to make some assumptions about the performance characteristics of a frequency domain optical storage system. We will follow the considerations of Ref [4], which contains a modeling study of such a system based upon single-photon PHB. A key performance parameter is the required data transfer rate. To be competitive with presently developed magnetic and optical disk storage systems we assume a read/write data rate of 30 ns per bit and shot-noise-limited reading with at least 26 dB wideband signal-to-noise ratio. Further, the medium area accessed by the laser beam must be small. A  $10\mu\text{m}$  diameter laser spot corresponds to  $10^6$  spatial locations per square centimeter, which, with the use of the frequency domain, yields an attractive total storage density of  $10^9 - 10^{10}$  bits/cm<sup>2</sup>. The photodetector is characterized by its quantum

efficiency  $\eta_Q$ . We assume that a transmission measurement technique is used for the data readout. Data are encoded in the frequency domain by the presence or absence of narrow spectral holes, each of which represents a characteristic absorption change  $\Delta\alpha$ , where  $\alpha = \sigma_1 N_\omega$  is the absorption coefficient. Here,  $\sigma_1$  is the low temperature absorption cross section of the optical transition and  $N_\omega$  is the concentration of optically active centers that are within a homogeneous linewidth of the laser frequency  $\omega$ . If  $N_{\text{tot}}$  is the overall concentration of centers whose resonance frequencies are distributed over the inhomogeneously broadened transition,  $N_\omega = (\Delta\omega_h/\Delta\omega_i)N_{\text{tot}}$ . A useful quantity for modeling purposes is the effective hole-burning yield  $\eta_e$ , which is defined as the relative absorption change  $\eta_e = \Delta\alpha/\alpha$ , that is produced by persistent spectral hole-burning during the writing time of 30 ns. Due to the complex nature of gated PHB processes the hole burning yield depends critically on the specific properties of the PHB material as well as on the writing conditions, which have to be compatible with a practical storage system. It is obvious that it is desirable to optimize the writing conditions to produce the largest yield  $\eta_e$  possible.

For data readout it is essential to discriminate between the optical transmission associated with a spectral hole and the original lower transmission characterizing the absence of a hole. The difference between the two corresponding photocurrents defines the amplitude of the reading signal  $S$  that has to be detected,

$$S = e\eta_Q(P/\hbar\omega)\{\exp[-(1 - \eta_e)\sigma_1 N_\omega L] - \exp[-\sigma_1 N_\omega L]\}, \quad (1)$$

where  $P$  is the laser power impinging on the photodetector,  $\hbar\omega$  is the photon energy,  $L$  is the thickness of the storage medium, and  $e$  is the electronic charge. Assuming shot-noise-limited detection the rms noise current averaged over the reading time  $\tau_R$  is given by

$$N = \{e^2\eta_Q(P/\hbar\omega\tau_R)[\exp(-(1 - \eta_e)\sigma_1 N_\omega L) + \exp(-\sigma_1 N_\omega L)]\}^{1/2}. \quad (2)$$



Eqs. (1) and (2) define the achievable signal-to-noise (voltage) ratio,  $S/N$ . We take  $S/N = 20$  (or 26dB) in a read time of 30 ns as the minimum value required for a practical storage system. Most of the parameters in Eqs. (1) and (2) are determined by the storage material itself. Thus, Eqs. (1) and (2) can be used to identify suitable combinations of material parameters so that  $S/N \geq 20$ .

In principle, the signal-to-noise ratio can be improved by increasing the read power. However, there are obvious restrictions on the maximum laser power that can be used for data readout. The maximum usable read power is either determined by the specific laser device being used or by the requirement of avoiding excessive spectral broadening of the written holes during reading. Many mechanisms can influence the practical width of the encoded spectral holes, including the effective laser linewidth, the Fourier transform width of the 30 ns writing and reading pulses, power broadening, the upper state lifetime  $T_1$ , and the dephasing time  $T_2$ . A detailed consideration of all these effects is beyond of the scope of this paper. Certainly, materials with  $T_1$  and  $T_2$  much shorter than  $\tau_R$  are undesirable since the associated fast relaxation would lead to inappropriately wide spectral holes that waste frequency space. In order to rule out excessive power broadening during readout, we choose a phenomenological limit on the laser power focused on the storage medium:  $P \leq 2A\hbar\omega/\sigma_1\tau_R$ , where  $A = 7.9 \times 10^{-7} \text{ cm}^2$  is the laser beam area corresponding to a beam diameter of  $10\mu\text{m}$ . It is worth mentioning that the maximum practical hole width is ultimately limited by the requirement for sufficient storage capacity in the frequency domain. In materials with very large inhomogeneous linewidth, very short relaxation times and/or power-broadening effects during data access may be tolerable within certain limits. For materials with high saturation intensities and low cross sections and therefore large lifetimes  $T_1$ , we assume an instrument-limited read power of  $P = 10 \text{ mW}$ . This is a typical power level for GaAlAs semiconductor diode lasers, which are the most practical laser sources presently available for

optical storage applications. Finally, we take  $\hbar\omega = 1.55$  eV and  $\eta_Q \approx 1$  for a state-of-the-art solid state photodetector.

With these considerations it is now possible to identify suitable material parameters  $\sigma_1$ ,  $N_\omega$ ,  $L$ , and  $\eta_e$  which result in  $S/N \geq 20$  (compare Eqs. (1), (2)). The thickness  $L$  of the storage material and the concentration  $N_\omega$  of the active centers are not intrinsic material properties and can be controlled when fabricating the storage medium. Thus, it is meaningful to classify materials by a concentration-thickness product  $N_\omega L$ . However, well-defined spatial resolution is required when accessing the stored information with the laser beam. Therefore, the material thickness  $L$  should not exceed the depth of field (as given by the Rayleigh range) associated with the focused laser beam of  $10\mu\text{m}$  diameter, i.e.  $L \leq 100\mu\text{m}$ . Cross sections of optical transitions are known for many materials or can easily be estimated and measured. Identifying materials by suitable combinations of  $\sigma_1$  and  $N_\omega L$  using the hole burning yield  $\eta_e$  as a parameter is particularly helpful for establishing guidelines for the search of promising PHB materials, and this is done in Figure 1. Materials within the shown boundaries for particular values of  $\eta_e$  permit data readout with a signal-to-noise voltage ratio  $S/N \geq 20$  using the reading conditions described above.

It is illustrative to describe the physical meaning of the boundaries in Figure 1. For high cross sections  $\sigma_1$  and large values  $N_\omega L$  the optical absorption becomes very strong. Consequently little light power impinges on the photodetector and the signal-to-noise ratio is low. For high  $\sigma_1$  and low values  $N_\omega L$  the achievable signal-to-noise is limited by the laser power that induces saturation broadening of the spectral holes. For  $\sigma_1 \leq 10^{-15} \text{ cm}^2$  the available laser power of 10mW is lower than the saturation power and thus defines the  $S/N$  limit. At the top of the figure, undesirable interactions between the optically active centers, e.g. energy migration and cooperative excited-state quenching, can restrict the usable

concentration of centers; this limit is highly material dependent. With a maximum material thickness of  $L = 100\mu\text{m}$ ,  $N_{\omega}L$  is ultimately limited to  $\approx 10^{16}\text{ cm}^{-2}$  since the corresponding total concentration of centers  $N_{\text{tot}}$  approaches the density limit of solid-state materials. In the fabrication of materials containing optically active centers it is usually not a problem to choose very low concentrations. However, in many cases high concentrations of centers are difficult to prepare or are intolerable for physical reasons. The right axis of Figure 1 gives the center concentration  $N_{\omega}$  under the assumption that the maximum media thickness  $L = 100\mu\text{m}$  is used. The results of the present analysis clearly show that a suitable PHB material has to contain a center concentration in excess of  $10^{13}\text{ cm}^{-3}$ . In case of gated PHB, the achievable yield  $\eta_e$  depends through complex relationships on the microscopic processes involved in the photo-induced material transformation as well as the amount of write power available for the frequency selective excitation and the subsequent gating process. However, spectral broadening of the produced hole, either by saturation of the transition or by excessive hole burning, imposes a limit on yield  $\eta_e$ . For this analysis, it seems justified to restrict the hole burning yield to  $\eta_e \leq 0.1$ . The  $\sigma_1 - N_{\omega}L$  parameter space shrinks rapidly as  $\eta_e$  decreases. In order to successfully implement a frequency domain optical storage system based on gated mechanisms, it is critical to find a material that permits gated PHB with very high efficiency in the short writing times required for fast data transfer rates.

Figure 2 provides a somewhat different view by classifying suitable PHB materials through a plot of effective yield  $\eta_e$  versus cross section  $\sigma_1$  using  $N_{\omega}L$  as a parameter. Here a given material has an optimum value of  $\eta_e$  and a particular value of  $\sigma_1$ , which defines a point in the  $\eta_e - \sigma_1$  plane. The contours show the required value of  $N_{\omega}L$  that must be achieved in order to obtain reading  $S/N \geq 20$ . Outside the allowed regions, various physical considerations prevent a solution to the materials optimization problem. At the top of the figure, holes would be expected to broaden unacceptably due to excessive photochemistry. At the lower left,

values of  $N_{\omega}L$  are required that are so large as to produce intercenter interactions such as spectral diffusion. Finally, at the lower right the required reading powers are large enough for saturation power broadening.

The lowest reading power included in Figures 1 and 2 is used for  $\sigma_1 \approx 10^{-11} \text{ cm}^2$ .

Saturation broadening limits the read power for this cross section to

$P \leq 2A\hbar\omega/\sigma_1\tau_R = 1.3\mu\text{W}$ . It is important to mention that such low reading power can be sufficient to obtain shot noise limited detection with  $S/N \geq 20$  with state-of-the-art high bandwidth photodiodes. Typical noise equivalent input powers of such devices are  $10^{-13} \text{ W}/\sqrt{\text{Hz}}$  corresponding to 0.41 nW for 30 ns reading time.

Based on a straightforward signal-to-noise analysis for data readout, key properties of gated PHB materials suitable for optical storage applications have been identified in Figures (1) and (2). No specific assumptions on technological details of the storage system were made with the exception of requiring a fast data rate of approximately 30 ns/bit with read  $S/N \geq 20$  and a laser focus of  $10 \mu\text{m}$  diameter for defining the saturation power. The analysis focused on the reading part of the problem, with all details of the writing process included in the effective yield,  $\eta_e$ .

### III. Photon-Gated Hole-Burning

To date, gated PHB has been observed in three materials:  $\text{Sm}^{2+}:\text{BaClF}$  [5],  $\text{Sm}^{2+}:\text{CaF}_2$  [7], and carbazole in boric acid glass[6]. In these examples, the gating mechanism involves absorption of a second photon, hence the name, "photon-gated". Photon-gated PHB is attractive for optical storage applications since there are no fundamental technical barriers for implementing such a process in the design of a practical device. Figures 3 and 4 illustrate some general aspects of photon-gated PHB mechanisms. In the case of a three level system (Fig. 3)

the absorption of a photon with energy  $\hbar\omega_1$  results in frequency selective excitation of level 2. The level 2 lifetime  $\tau$  should not be so short as to produce unacceptably broad holes. Absorption of a second photon of energy  $\hbar\omega_2$  excites the center to level(s) 3. From level 3 the photoreaction occurs with microscopic quantum efficiency  $\eta$ , reducing the population of centers that are resonant at  $\omega_1$ . This is the general level scheme for the system  $\text{Sm}^{2+}:\text{BaClF}$ , in which  $N_{\omega}L \approx 10^{14}\text{cm}^{-2}$  and  $\sigma_1 \approx 10^{-18}\text{cm}^2$ . Note that the effective hole burning yield  $\eta_e$  used in the signal-to-noise analysis above depends on the absorption cross sections  $\sigma_1$  and  $\sigma_2$  as well as on the microscopic reaction efficiency  $\eta$  (compare Fig. 3). There is no fundamental need for a frequency-selective narrow band transition from level 2 to level 3. When the excitation energy  $\hbar\omega_2$  exceeds a certain threshold value, the photoreaction is initiated, causing the formation of a persistent spectral hole. In cases where the lifetime  $\tau$  of level 2 is much larger than the data access time  $\tau_R$ , the storage material does not have to be exposed with both photon energies simultaneously. By rapid variation of  $\hbar\omega_1$  selected centers can be excited to level 2 with characteristic lifetime  $\tau$ . After a time period shorter than or comparable to  $\tau$ , irradiation with  $\hbar\omega_2$  induces the photoreaction. The exposure time of this second step can be rather long without loss of writing speed.

In principle, systems with  $\omega_1 = \omega_2$  can exhibit gated PHB in the sense that the hole burning yield is nonlinear with laser intensity. However, the requirement of non-destructive reading makes it desirable to use systems with  $\omega_1 \neq \omega_2$  permitting complete decoupling of reading and writing processes. For efficient gated hole burning it is advantageous to choose centers where  $\omega_1$  is not absorbed from the excited state 2 (region a) and where  $\omega_2$  does not cause excitations from the ground state (region b). Suitable values for  $\sigma_1$  have been determined by the analysis in Section II. The cross section  $\sigma_2$  should certainly be large to assure efficient utilization of the available gating light and permit high hole-burning yield  $\eta_e$ . Thus in certain instances,

narrow band levels 3 involving transitions with high peak cross sections may be preferable to a continuum such as a conduction band.

Figure 4 illustrates photon-gated PHB involving a 4-level system. This type of level scheme is appropriate for organic molecules or color centers that have states with different spin multiplicity. For the case of carbazole in boric acid glass,  $N_{\omega}L \approx 10^{13} \text{ cm}^{-2}$  and  $\sigma_1 \approx 5 \times 10^{-12} \text{ cm}^2$ . After the initial frequency selective excitation  $1 \rightarrow 2$ , the system relaxes to the intermediate state  $i$  by intersystem crossing, for example. Subsequent absorption of a second photon by the  $i \rightarrow 3$  transition induces persistent spectral hole burning. For efficient gated PHB the  $2 \rightarrow i$  relaxation rate  $\Gamma_i$  should be as large as possible consistent with the requirement that the lifetime of level 2 not be too short. Further, a long intermediate state lifetime  $\tau_i$  would be advantageous in achieving a large population in level  $i$  for excitation to level 3. It is evident that absorptions  $2 \rightarrow (a)$ ,  $1 \rightarrow (b)$ , and  $i \rightarrow (c)$  involving photons of frequency  $\omega_1$ ,  $\omega_2$ , and  $\omega_1$ , respectively, should not be large (compare Fig. 3). Of course, the microscopic yield  $\eta$  should be as large as possible for efficient photon gating.

A further constraint for promising materials might be the availability of practical light sources to provide the needed frequencies  $\omega_1$  and  $\omega_2$ . A narrow linewidth, rapidly tunable laser such as a GaAlAs or other semiconductor diode laser is essential for frequency selective excitation at  $\omega_1$ . The much less stringent requirements for the gating light can, in principle, permit use of incoherent sources such as (flash) lamps, light emitting diodes, or super luminescent diodes.

None of the recently discovered materials that exhibit photon-gated PHB fulfill all the requirements for a successful frequency domain optical storage medium. A final evaluation of these materials will require determination of the hole-burning yield  $\eta_e$  that can be achieved during the 30 ns writing time. Independent of the yield  $\eta_e$ , the low cross section and limited

concentration of  $\text{Sm}^{2+}$  in BaClF would require very high read powers on the order of 1 W to achieve  $S/N \geq 20$ . For the material composed of carbazole in boric acid glass, practical tunable lasers in the near ultraviolet would be needed.

#### IV. Conclusions

In conclusion, several guidelines in the search for gated PHB materials suited for frequency domain optical data storage have been established by identifying appropriate values for important material properties such as concentration of active centers, absorption cross section, and effective hole-burning yield. The results of this analysis permit evaluation of the suitability of newly discovered materials exhibiting gated PHB. The specific case of gated PHB involving a photon-induced gating mechanism has been discussed and desirable properties of two types of photon-gated mechanisms have been described. Innovation in the development of gated PHB materials will be crucial for the design of a practical high-density storage system based upon persistent spectral hole burning.

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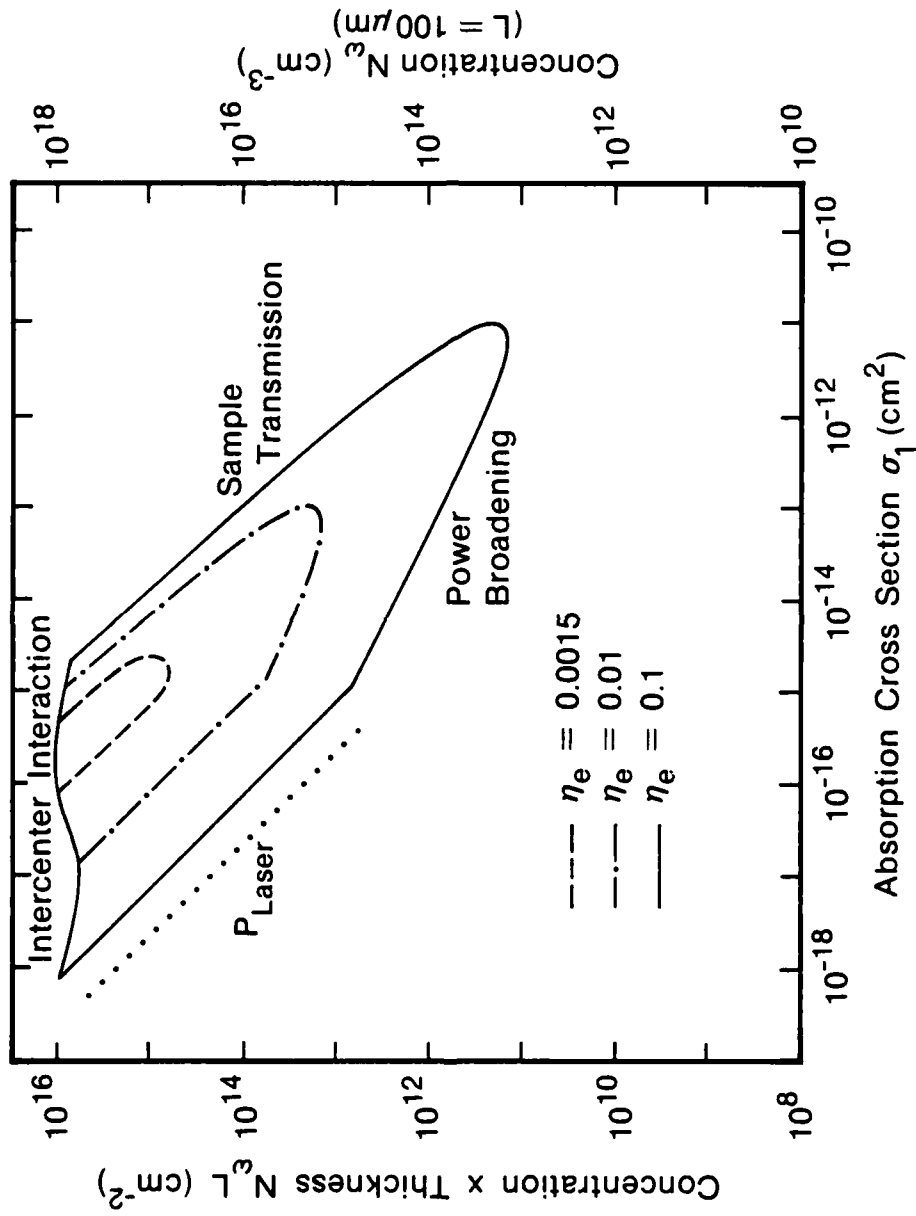


Figure 1.

Materials constraints for gated PHB materials in order to achieve practical S/N ratios. The various regions and symbols are defined in the text. To illustrate the effect of increased laser power, the dotted line shows the  $\eta_e=0.1$  boundary for 100 mW reading power.

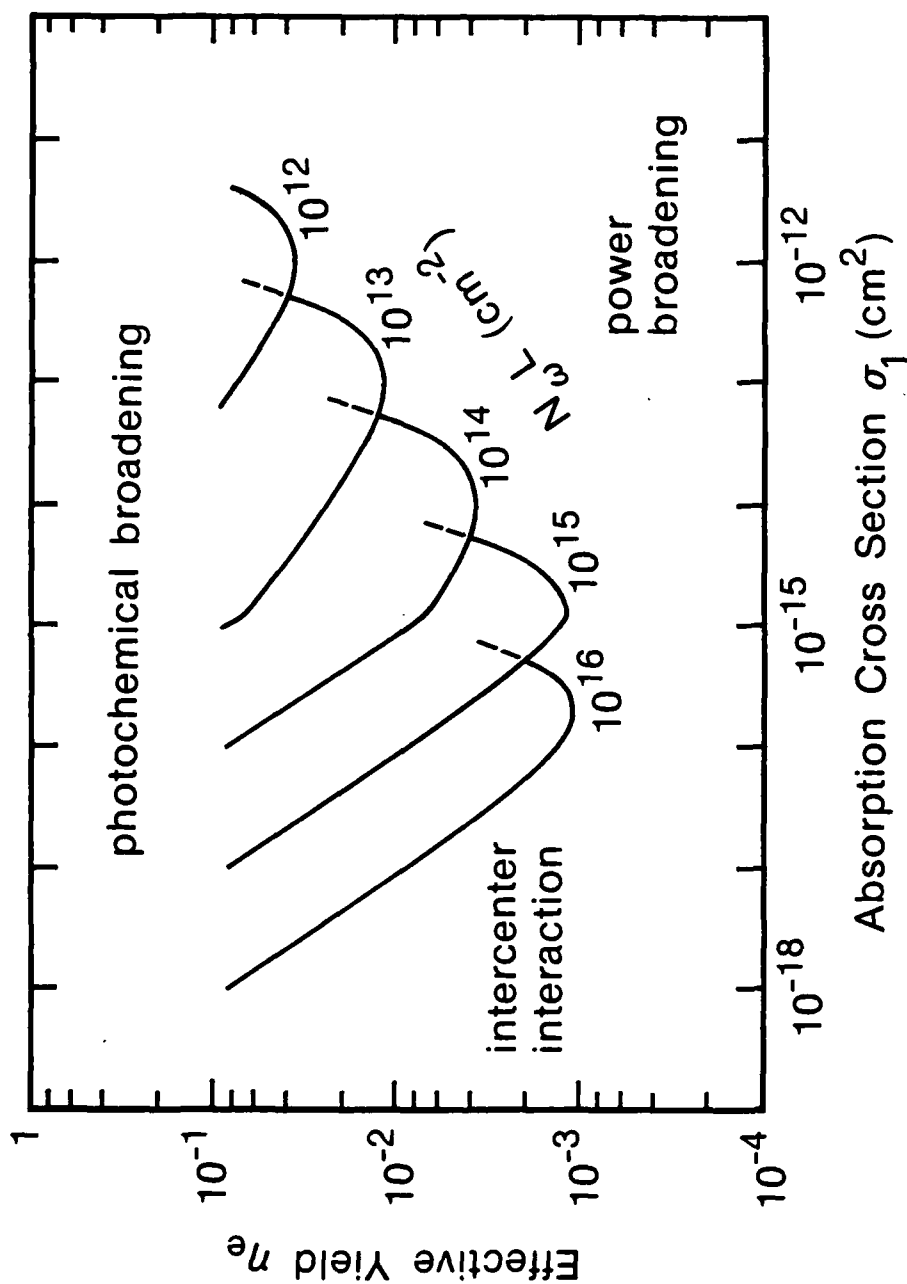


Figure 2.

Materials constraints presented as contours of the required density-thickness product  $N_{\omega}L$  in the  $\eta_e - \sigma_1$  plane. Three forbidden regions are indicated as described in the text.

### 3-Level Gated Material

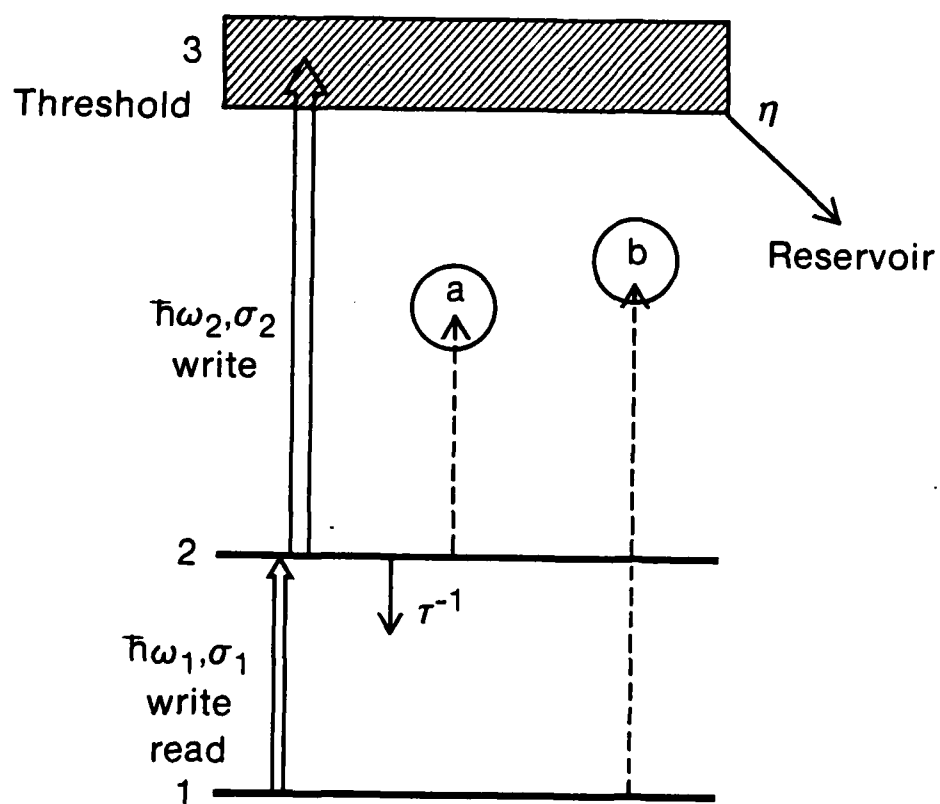


Figure 3.

General level structure of a three-level photon-gated PHB mechanism. The absorption should be small in the regions labeled (a) and (b).

### 4-Level Gated Material

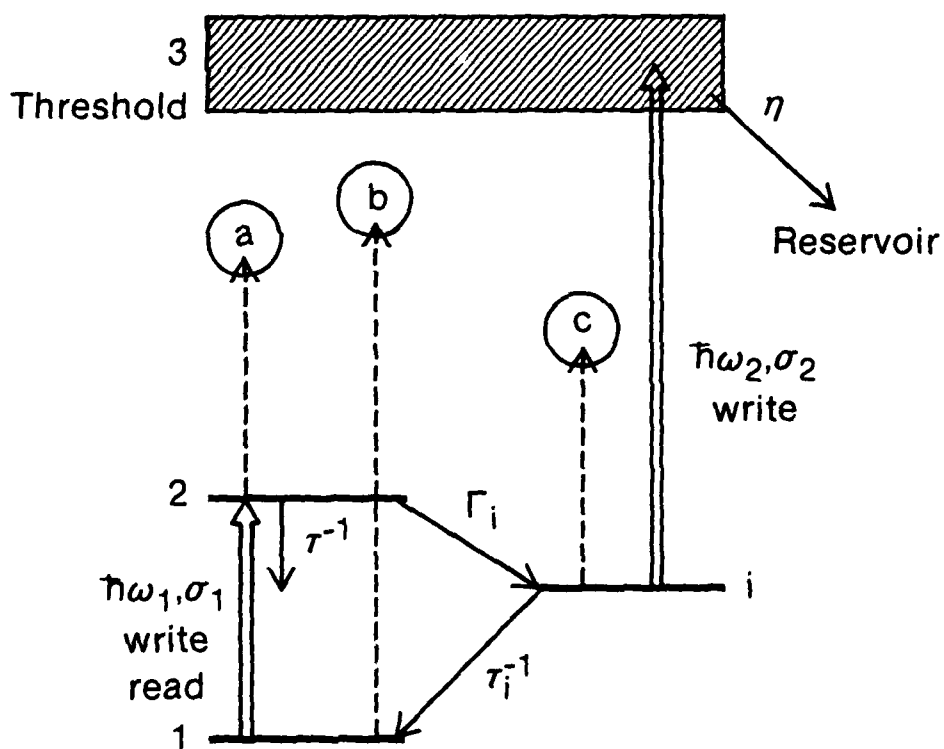


Figure 4.

General level structure of a four-level photon-gated PHB mechanism. The absorption should be small in the regions labeled (a), (b), and (c).

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